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MEASURING AND CORRELATING NEUTRON EXPOSURE
AND DAMAGE IN GRAPHITE

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MEASURING AND CORRELATING NEUTRON EXPOSURE AND DAMAGE IN GRAPHITE

INTRODUCTION

This report describes methods for predicting service performance of graphite when the test-reactor environment differs from actual or anticipated environment. Although the application here is to graphite, the methods are applicable to other materials that are damaged by atomic displacements caused by reaction irradiation.

Correct interpretation of dosimetry data is critical to the determination of neutron exposure. The threshold monitors (Ni, Fe, Ti), which are used to monitor exposures in damage experiments, measure only about 10% of the neutrons which damage graphite. To extrapolate the activation data over an energy range sufficiently great to correlate damage, a neutron spectrum must be assumed or calculated. Unless the spectrum chosen is accurate, gross errors in exposure estimates will result.

Appropriate constants for the reduction of data in individual experiments are listed which will permit the experimenter to calculate neutron exposures with the best techniques currently available.

SUMMARY

A procedure is described for measuring neutron exposure to graphite and for correlating radiation-damage data from different types of reactors. The fast-neutron spectrum is calculated and is, in turn, used to calculate the activation in threshold monitors such as nickel and iron. Nickel and iron are activated by neutrons with energies greater than about 3 Mev, so an effective cross section is computed to calculate exposures to neutrons with energies greater than 0.18 Mev. With these effective cross sections, activation monitors at several reactors yield exposures in agreement to $\pm 10\%$.

The ratio of gross atomic-displacement rate to the flux greater than 0.18 Mev was constant within 10% in several neutron spectra. If the magnitude of radiation-induced changes is proportional to displacement production, data from various reactors can be related by the exposure unit, nvt ($E > 0.18$ Mev).

Conversion units from megawatt day per adjacent ton to integrated fast flux have been tabulated. Results are 1.19×10^{17} nvt/(Mwd/At) in the Hanford K-Reactor and 0.96×10^{17} nvt/(Mwd/At) in the Hanford C-Reactor.

IRRADIATION FACILITIES

The irradiation facilities that were analyzed included test positions in the Hanford Reactors, the General Electric Test Reactor (GETR), and the Engineering Test Reactor (ETR). The Hanford N-Reactor lattice was also analyzed in detail since the test data are used to predict property change in the N-Reactor moderator.

Test holes in C- and K-Reactors have been used extensively for graphite irradiations for many years. These are square holes which run through the graphite moderator normal to and between fuel channels in the reactor. The K-Reactor lattice and the representation used in the analyses are illustrated in Figure 1. C-Reactor is very similar except that the lattice pitch is $8 \frac{3}{8}$ in. Both reactors were analyzed in the same manner and cell-boundary spectra were used to obtain dosimetry and damage parameters for the facilities.

FIGURE 1
K-Reactor Lattice
and Process Tube Details

Graphite irradiations are conducted in several positions in the northwest quadrant of the ETR (Figure 2) including E-5, F-6, and G-6. For the cases analyzed, "hot capsules" were located in E-5 and G-6 and a graphite creep test in F-6.

Positions in the GETR core used for graphite irradiations are illustrated in Figure 3.

The N-Reactor lattice analysis yielded parameters for each 1/2 in. square in the lattice; however, data cited in this report represent an average for the area shaded in Figure 4 since this region is of principal interest.

FIGURE 2

ETR Core
(The Northwest quadrant
used in the flux calculations
lies inside the heavy lines.)

CALCULATION PROCEDURE

Neutron spectra were calculated with one of two codes, depending upon the geometry of the facility. Those with radial symmetry were calculated with PROGRAM S (GE-HAPO-S-12),⁽¹⁾ and the others were calculated

FIGURE 3

GETR Core
as Represented
for the Analysis

FIGURE 4

N-Reactor Lattice and Area
Used in Transport-Theory Calculations

with 2DXY.⁽²⁾ Both programs are multigroup, multiregion, transport-theory codes. All problems were solved with 16 energy groups in the range 0.18 to 10 Mev. Cases run with 2DXY were computed with two additional groups spanning the energy range below 0.18 Mev. Cases run with PROGRAM S utilized 17 energy groups below 0.18 Mev. Exact group structures are shown in Table I.

Flux-averaged cross sections for reactor materials used in the transport-theory analyses were generated by the GAM⁽³⁾ code. Each reactor or lattice was homogenized and a spectrum calculated in a zero dimension analysis. These spectra were then used to obtain group averaged cross sections for reactor materials.

TABLE I
GROUP STRUCTURE AND GROUP AVERAGED CROSS SECTIONS

Group	E_L , ev [*]	U_L **	σ , barns		Displacement Production in Carbon ⁽⁵⁾
			Ni ⁵⁸ (n, p) Co ⁵⁸ ⁽⁴⁾	Fe ⁵⁴ (n, p) Mn ⁵⁴ ⁽⁴⁾	
1	7.79×10^6	0.25	0.651	0.310	2378
2	6.07×10^6	0.50	0.622	0.270	1739
3	4.72×10^6	0.75	0.552	0.250	1646
4	3.68×10^6	1.00	0.349	0.220	2691
5	2.87×10^6	1.25	0.200	0.190	2964
6	2.23×10^6	1.50	0.116	0.130	2653
7	1.74×10^6	1.75	0.048	0.050	2716
8	1.35×10^6	2.00	0.015	0.005	2745
9	1.05×10^6	2.25	0.004	0	3205
10	8.21×10^5	2.50	0		3078
11	6.39×10^5	2.75			3063
12	4.98×10^5	3.00			2970
13	3.88×10^5	3.25			2817
14	3.02×10^5	3.50			2573
15	2.35×10^5	3.75			2321
16	1.83×10^5	4.00			2030
17	1.11×10^5	4.50			1496
18	6.74×10^4	5.00			1060
19	4.09×10^4	5.50			620
20	2.48×10^4	6.00			380
21	1.50×10^4	6.50			240
22	9.12×10^3	7.00			140
23	5.53×10^3	7.50			70
24	2.04×10^3	8.50			30
25	454.	10.1			6
26	130.	11.25			0
27	78.9	11.75			
28	47.9	12.25			
29	29.0	12.75			
30	13.7	13.50			
31	5.04	14.50			
32	0.414	17.0			
33	0	∞			

* E_L Energy at lower limit of group

** U_L Lethargy at lower energy limit of group

The number of spatial zones utilized in the calculations depended upon the problem and the type of studies in the facility. Thirty-five regions were calculated in the Hanford K- and C-Reactor lattices by PROGRAM S. The GETR and ETR cores and the N-Reactor lattice were analyzed with 2DXY. The GETR core was represented with 441 zones, each 1 by 1 in.; the ETR core with 361 1 x 1 in. zones; and the N-Reactor lattice with 248 1/2 by 1/2 in. zones. Materials were smeared within the zones. Each 3 by 3 irradiation position in the test reactors had 9 subdivisions, thus retaining a high degree of accuracy and detail. All calculations were made on an IBM 7090 computer with approximately 8 to 10 hr required for an analysis of each reactor or lattice.

Spectra calculated for C-Reactor and for the ETR are shown in Figure 5 to illustrate differences in typical spectra in the two types of reactors.

FIGURE 5

Flux Spectra in ETR Test Facility
and a Graphite Moderated Reactor

NEUTRON DOSIMETRY

Neutron exposures for irradiation tests should be calculated from activation of monitors with effective cross sections appropriate for each irradiation facility. Fast-neutron exposures are monitored in irradiation tests by iron and nickel wires or foils. Radiochemical analysis of the products of the $\text{Fe}^{54} (n, p) \text{Mn}^{54}$ and $\text{Ni}^{58} (n, p) \text{Co}^{58}$ reactions provide the basis for calculating the exposure. A major problem arises because the flux must be calculated over the energy range 0.18 to 10 Mev to correlate graphite damage, yet the monitors are activated only by neutrons with energies above 3 Mev. Thus, an energy range below the activation range must be included through effective cross sections:⁽⁶⁾

$$\sigma_{\text{eff}} = \frac{\int_0^{10 \text{ Mev}} \sigma(E) \varphi(E) dE}{\int_{E_0}^{10 \text{ Mev}} \varphi(E) dE}$$

where σ_{eff} is the effective cross section for energies greater than E_0

$\sigma(E)$ is the differential cross section for the nuclear reaction

$\varphi(E)$ is the flux spectrum

E_0 is the base energy for the effective cross section.

The differential cross-section data in Table I were used to calculate the effective cross sections presented in Table II. The variations between the effective cross sections in the test reactors and those in the Hanford reactors are due to spectral differences in the energy range, 0.18 to 3.0 Mev, that does not activate the monitors but that does cause the most radiation damage. The need for detailed analysis is apparent for accuracy in exposure determinations.

TABLE II
EFFECTIVE CROSS SECTIONS AND DISPLACEMENT PRODUCTION

Reactor or Irradiation Facility	$\phi^*(>0.18 \text{ Mev})$ Mwd/At $\times 10^{17}$	$\phi(>0.18 \text{ Mev})$ $\phi > 1 \text{ Mev}$	$\sigma_{\text{eff}} (\phi > 0.18 \text{ Mev})$ mbarns		Displacement Production $\phi > 0.18 \text{ Mev}$
			Fe ⁵⁴ (n, p) Mn ⁵⁴	Ni ⁵⁸ (n, p) Co ⁵⁸	
N (See Figure 4)	1.88	2.54			3283
K (Hot Test Hole)	1.19	2.60	27.7	38.1	3439
C (Hot Test Hole)	0.95	2.64	27.9	38.6	3460
GETR					
D-7		2.00	40.2	62.4	3019
E-7		1.82	51.4	84.3	3017
F-7		1.98	40.3	62.6	3014
ETR					
F-6		1.83	49.3	76.6	3022
G-6					
NW		1.84	50.0	77.7	2961
NE		1.80	52.6	81.9	2929
SE		1.78	48.2	75.3	3017
SW		1.89	54.0	84.6	2939
E-5					
NW		2.23	26.6	39.6	3172
NE		1.95	43.1	66.9	3005
SE		1.86	40.4	61.9	3032
SW		1.97	46.1	70.8	2973

$$* \phi = \int_{E_0}^{\infty} \varphi(E) dE$$

The accuracy of the effective cross sections, however, is no better than the accuracy of the differential cross sections. This is presently a severe problem with iron because of the very limited number of cross-section measurements at energies below 10 Mev. It has been necessary to postulate a differential cross section curve to develop effective cross sections.⁽⁴⁾ Although application of the results yields exposures in good agreement with those determined with nickel monitors (within 15%), better agreement can be expected as more cross-section measurements are made.

Effective cross sections have been calculated to give exposures in units that have been used to correlate graphite damage, i.e., $E > 0.18$ Mev. The values that have been presented can be converted to calculate $\Phi(E > 1.0 \text{ Mev})$:

$$\sigma_{\text{eff}} [\Phi(E > 1.0 \text{ Mev})] = \sigma_{\text{eff}} [\Phi_{\text{eff}} [\Phi(E > 0.18 \text{ Mev})] \frac{\Phi(E > 0.18 \text{ Mev})}{\Phi(E > 1.0 \text{ Mev})}.$$

The flux ratio for each irradiation facility is presented in Table II.

The effective cross sections presented in this report have been used to calculate exposures for irradiation experiments in the ETR,⁽⁷⁾ and for the first time agreement has been obtained between exposures calculated from nickel and from iron monitors. These data (Table III) show agreement within about 15% in most of the experiments. This agreement is of particular significance since it enables one to place more reliance on exposures calculated from iron monitors.

TABLE III
EXPOSURES Φ ($E > 0.18 \text{ Mev}$) CALCULATED FROM MONITORS
IRRADIATED IN THE G-6 FACILITY IN THE ETR

Capsule	Distance from Top of Core, in.	$\Phi \times 10^{-21}$ ($E > 0.18 \text{ Mev}$)	
		Fe	Ni
162	6	1.02	0.91
163	12	1.18	1.30
164	18	1.31	1.43
165	24	1.16	1.19
166	30	0.76	0.82
176	6	0.51	0.98
177	12	1.60	1.53
178	18	--	1.00
179	24	1.38	1.20
180	30	0.86	0.84

DAMAGE NORMALIZATION

Correlation of radiation-effects data is often very difficult because of the influence of spectra on damage accumulation. Theory⁽⁸⁾ has indicated

that, if exposures for graphite are measured in units of neutrons per flux integrated for energies greater than 0.065 Mev, spectral differences are normalized. Recently, exposures for graphite irradiations have been expressed as flux greater than 0.18 Mev, nvt ($E > 0.18$ Mev). Displacement rates per integrated flux ($E > 0.18$ Mev) have been calculated for each facility according to the de Halas model⁽⁵⁾ and are presented in Table II. These ratios are constant to within $\pm 10\%$, demonstrating that this normalization base nearly corrects for spectral variation and is quite accurate.

Further refinement is readily attained by correlating exposures between facilities with these ratios. As an example, the ratio of displacements to integrated flux for N-Reactor is 3283 and that for the D-7 facility in the GETR is 3019; thus, an integrated exposure of 10^{21} nvt ($E > 0.18$) would cause $\frac{3283}{3019}$ or 1.09 times the number of carbon-atom displacements in N-Reactor than in D-7. An "effectiveness" index for the facilities is thereby established.

The effects of spectral variation on damage accumulation in graphite, and the need for detailed spectral determinations are illustrated in Figure 6. The fraction of total displacement production is plotted against neutron lethargy for the GETR, the C-Reactor spectrum, and for an unmoderated Watt fission spectrum. Damage accumulation is quite different. Neutrons with energy above 1 Mev cause approximately 90% of the total damage in a fission spectrum but only about 35% of the total damage in the C-Reactor spectrum. The lower energy flux component, $0.1 \text{ Mev} > E > 0.005$, causes 20% of the damage in the relatively well moderated C-Reactor spectrum but less than 5% in the GETR. Finally, neutrons with energy greater than 3 Mev, which produce activation in monitors, cause less than 5% of the total damage in the three spectra. Detailed knowledge of neutron spectra are, therefore, essential if accurate damage studies are to be made.

CONVERSION OF EXPOSURE DATA TO REACTOR ENERGY PRODUCTION

Conversion between fast neutron exposure units such as nvt ($E > 0.18$ Mev) and units of energy production in fuel adjacent to experiments is of interest because a great deal of data have been presented in each system.

FIGURE 6

Displacement Production **Modes** in Several Neutron Spectra

Units of megawatt days per adjacent ton of fuel are used to express exposure in terms of local energy generation.

Conversion factors have been calculated, according to the method derived in the Appendix, that give the ratio of flux ($E > 0.18$ Mev) to Mwd/At, in three Hanford reactors and are presented in Table II. Values cited for C- and K-Reactors are applicable to hot test hole facilities within the fueled region of the reactor. The value for N-Reactor was calculated to apply to the graphite in a tube block directly under the tube.

Significant differences in the conversion ratios may be noted among the three reactors, C, K, and N. Although all are graphite moderated and somewhat similar in core design, a factor of 2 is obtained between

C and N, and a 25% difference between C and K. These differences are primarily due to differences in the fuel-to-moderator ratios in the reactors. However, the most striking fact is that if damage estimates are made on the basis of Mwd/At exposures, errors of 100% would result. These errors would occur because reactor power is proportional to the thermal flux, whereas damage is dependent on the fast flux. Ratios of fast-to-thermal flux are considerably different among facilities and can vary significantly during an irradiation cycle in any particular facility. Since variation exists, it is necessary to specify the reactor upon which the Mwd/At normalization is based. This is not necessary for exposures which are properly normalized in terms of integrated fast flux; hence, the latter unit is more accurate and convenient.

The conversion ratios presented in this report were obtained from reactor analysis computations. Since these ratios are the basis for exposure normalization for a great deal of irradiation test data, it was necessary to check the ratios with experimental observations as closely as possible. Ideally one would experimentally measure the flux ($\Phi E > 0.18$ Mev) in an irradiation facility and power generation in the adjacent fueled region. Power generation can be measured with reasonable accuracy, but fast flux ($\Phi E > 0.18$ Mev) must be determined through the use of effective cross sections. A completely independent check is not possible. The most pertinent check is a determination of the ratio from monitor data reduced with the effective cross sections calculated for the position and local power generation measurements.

Morgan⁽⁹⁾ calculated the conversion ratios in Table II to check the theoretical analysis using the effective cross sections for nickel presented in Table I to calculate the fast flux ($\Phi E > 0.18$ Mev); tube-power data were obtained from reactor operational records. The local power generation (Mwd/At) was calculated by assuming a chopped cosine distribution of power along the tubes adjacent to the irradiation position. Flux-to-power ratios were calculated with monitor data from ten experiments irradiated in a hot test hole in K-Reactor. Ratios ranged between 1.17×10^{17} nvt/(Mwd/At) and 1.37×10^{17} nvt/(Mwd/At). The average of these data was $1.24 \pm 0.09 \times 10^{17}$ nvt/(Mwd/At) which agrees very well with

the theoretical value of 1.19×10^{17} . Similar experiments in a C-Reactor hot test hole yielded ratios that ranged from 0.775×10^{17} to 1.14×10^{17} and averaged 0.959×10^{17} ; this may be compared to the theoretical value of 0.953×10^{17} . The agreement between theoretical calculations and the experimental data is very good especially in this type of facility. These ratios can, therefore, be used with confidence in resolving data generated in the past in these facilities.

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APPENDIX

DERIVATION OF EXPOSURE UNIT CONVERSION

The ratio between integrated fast flux in a test facility and local power generation is basically obtained from a transport theory analysis of the lattice. This analysis gives the ratio of the fission rate in the fueled zone to the flux in the zone used for irradiation testing. This ratio is then multiplied by constants to obtain the conventional units of megawatt days per adjacent ton.

When computing these ratios a complication is introduced since the transport theory analyses were made in either one or two dimensions with the inherent assumption that the system was invariant in the other dimensions. These were satisfactory approximations in the Hanford reactors.

The equation which relates the fast flux in one zone to power generation in another is derived for use within transport-theory analyses is as follows.

The flux integrated over the energy range that correlates radiation damage, and over the zone in which radiation damage occurs is:

$$\phi dt = \frac{\int_{Z_1}^{Z_2} \int_{Y_1}^{Y_2} \int_{X_1}^{X_2} \int_{E_0}^{D_0} \phi(E, X, Y) dE dX dY dZ}{\int_{Z_1}^{Z_2} \int_{Y_1}^{Y_2} \int_{X_1}^{X_2} dX dY dZ} \quad (1)$$

The fission rate per atom of fuel (uranium) in the adjacent fuel is:

$$\frac{dU_f}{U} = \frac{\int_{Z_1}^{Z_2} \int_{Y_0}^{Y'} \int_{X_0}^{X'} \int_0^{\infty} \phi(E, X, Y) \Sigma_f(E, X, Y) dE dX dY dZ}{\int_{Z_1}^{Z_2} \int_{Y_0}^{Y'} \int_{X_0}^{X'} N_u(X, Y) dX dY dZ} \quad (2)$$

In Equations 1 and 2

Z_1 and Z_2 are the limits of integration in the Z domain and are the same in each integration.

X_1 , X_2 , Y_1 , and Y_2 are the spatial limits of the fuel.

X_0 , X' , Y_0 , and Y' are the spatial limits of the irradiation testing facility.

E_0 is the lower energy limit used to correlate radiation damage in material.

$\frac{dU_f}{U}$ is the fission rate of fuel (uranium) per second per uranium atom.

Σ_f is the macroscopic fission cross section = $\sigma_f N_u$.

N_u is the concentration of uranium atoms per unit volume.

The elements in Equations 1 and 2 are obtained from the transport theory analyses. Thus, if Equation 1 is divided by Equation 2 one obtains:

$\frac{\phi dt}{\left(\frac{dU_f}{U}\right)}$ which is the integrated flux per fission. To convert this to local

power generation the constant relating power per fission and one defining average fuel loading are introduced. Thus

$$\frac{\phi}{P/M} = \frac{\phi dt A \bar{\rho}_u}{\left(\frac{dU_f}{U}\right) N_u B} = \frac{\phi (E > E_0)}{P/M} [\phi (Mwd/At)]$$

where $A = 3.70 \times 10^{-22} \frac{Mwd}{\text{fission}}$, based on $\frac{200 \text{ Mev}}{\text{fission}}$

P = power

M = mass of uranium

N_u = uranium atoms/cm³

$\bar{\rho}_u$ = uranium density averaged over fuel channel (g/cm³)

B = g/ton

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